TITLE: Minimizing NOx Emissions from Multi-Burner Coal-Fired Boilers

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ABSTRACT

OBJECTIVE

The focus of this program is to provide insight into the formation and minimization of NOx in multi-burner arrays, such as those that would be found in a typical utility boiler. Most detailed studies are performed in single-burner test facilities, and may not capture significant burner-to-burner interactions that could influence NOx emissions.

Our approach is to investigate such interactions by a combination of single and multiple burner experiments in a pilot-scale coal-fired test facility at the University of Utah, and by the use of computational combustion simulations to provide insight into the experimental results and to evaluate full-scale utility boilers.

The program is broken into four main tasks:

1- Fundamental studies on nitrogen release from coal. These studies will be used to enhance the predictive capabilities of the combustion simulations.
2- Comprehensive modeling of burner arrays.
3- Pilot-scale optimization of multi-burner arrays.
4- Technology transfer.

ACCOMPLISHMENTS TO DATE

Significant progress has been made on the fundamental studies of N release from coal, as such understanding is key to subsequent tasks in the program. Two different experimental programs are underway to obtain this information: Flat Flame Burner studies at BYU under the direction of Prof. Tom Fletcher; and drop-tube studies at the University of Utah under the direction of Prof. Adel Sarofim.

Experimental studies of nitrogen release during secondary pyrolysis include pyrolyzing four coals in a flat flame reactor in the BYU combustion laboratory. Temperature, residence time and coal rank are chosen as the test variables in the experiments. Experiments were made at temperatures ranging from 1160 K to 1850 K and at residence times from 18 ms to 100 ms under fuel-rich conditions (i.e., 0% post-flame O_2). The four coals used are Illinois #6, Utah bituminous, Wyodak subbituminous coal, and Knife River

lignite. Elemental analyses (C, H, N, S) were made on the corresponding parent coal, tar, soot and char. Estimation of the daf mass release was based on a tracer technique which uses relatively small amounts of Ti, Al and Si on the char/coal pair by ICP (Inductively Coupled Plasma). An FTIR spectrometer was used with a 10-m multi-reflection gas cell to measure gas species concentrations, including HCN, NH_3, and hydrocarbon species. About 80 experiments have been performed to date.

Analysis of the data is currently underway. The lignites exhibit HCN and NH_3 release at low temperatures, while the higher rank coals show no NH_3 at the low temperatures. As temperature increases, the HCN and NH_3 yields both increase for all coals. The low temperature HCN yield seems to be correlated with the yield of C_2H_2 (acetylene) from each coal. The nitrogen in the tar/soot decreases with increased temperature for the range of temperatures examined. ^{13}C NMR analyses were made on certain tar/soot samples to determine the chemical structure changes of tar during secondary reactions. In addition, soot samples from two model aromatic compounds (bi-phenyl and pyrene) were obtained in the flat flame ractor, and are currently being analyzed by a variety of ^{13}C NMR techniques.

A comprehensive literature review of the char-nitrogen conversion process allowed the investigators to identify char nitrogen as an important source of NO and N_2O emissions during low NOx coal combustion. This review was accepted for publication in Progress in Energy and Combustion Science[1].

A complementary result of the review was the identification of the difficulty of existing 3-D modeling codes to predict the evolution of char-N during combustion. This lead the investigators to develop a simple model with a single NO formation step during char nitrogen oxidation and a first order reaction between NO and char. This model captures the main characteristics of the NO-char kinetic reactions at pulverized coal combustion conditions; in particular, the very significant decrease in the apparent conversion of char nitrogen to NO with increasing ambient NO concentrations. The model was calibrated with experimental data obtained for char combustion for pulverized fuel in a controlled atmosphere in a bench-scale combustor[2]. *Ab Initio* chemistry calculations were also carried out to investigate specific pathways in the char/NO interactions, to determine controlling reaction sequences and thus allow modification of the current single particle model.

Incorporation of the model in a CFD code provides interesting insights on the role of char nitrogen on NO formation in pulverized coal boilers[3]. Tracing of over a thousand particles as a post-processor showed that individual particles had an apparent char nitrogen conversion to NO which ranged from slightly negative to ninety percent. The average conversion was in the range of ten percent, satisfyingly close to values that had been found to empirically fit data on a series of coals fired in a bench-scale furnace[4].

Two additional tasks have been identified in order to strengthen the current char-N model. Task number one involves an experimental study of the fate of char-N at pulverized coal combustion conditions in a controlled atmosphere. These experiments will be conducted in a laminar flow drop tube reactor that was set up during 1999 in the combustion laboratory at the University of Utah in order to perform this project. The experimental study will complement previous research in the laboratory[2] by diminishing the influence of neighboring particles on the reduction of NO. The second task will implement the single particle model directly into Glacier, thus allowing a dynamic interaction of the particle model with the CFD code and improving the postprocessor technique used in the last publication[3].

During this year, the multi-burner firing configuration has been installed on the L1500 pilot-scale furnace and preliminary experimental data has been obtained. The data have highlighted some difficulties in obtaining flame characteristics and NOx emissions similar to those obtained with a single burner. The pilot-scale testing will continue through the summer months, focusing on detailing the differences between single- and multi-burner operation and optimizing the system to achieve minimum NOx emissions.

REFERENCES

1 - Molina, A., Eddings, E.G., Pershing, D.W. and Sarofim, A.F. “Char Nitrogen Conversion: Implications for Emissions from Coal-Fired Utility Boilers,” Prog. Energy Combust. Sci., *accepted for publication*, 2000.
2 - Spinti, J. “An experimental study of the fate of char-nitrogen in pulverized coal flames,” Ph.D. Thesis, University of Utah, 1997.
3 - Sarofim, A.F., Pershing, D.W., Eddings, E.G. and Molina, A. “NO-Char Kinetics: Implementation to NOx Emissions,” Mediterranean Combustion Symposium, Antalya, Turkey, 1999.
4 - Pershing, D.W. and Wendt, J. “Pulverized Coal Combustion: The Influence of Flame Temperature and Coal Composition on Thermal and Fuel Nox,” Sixteenth Symposium (International) on Combustion/The Combustion Institute 1976, 389.

ARTICLES, PRESENTATIONS AND STUDENTS

JOURNAL ARTICLES (peer reviewed)

Molina, A., Eddings, E.G., Pershing, D.W. and Sarofim, A.F. “Char Nitrogen Conversion: Implications for Emissions from Coal-Fired Utility Boilers,” Prog. Energy Combust. Sci., *accepted for publication*, 2000.

CONFERENCE PRESENTATIONS

E. G. Eddings, R. Okerlund, D. W. Pershing, M. P. Heap, R. Beittel, R. Lisauskas, “Optimization of a Low NOx Firing System for High-Temperature Slagging Systems,” poster presentation at the 28th Symposium (International) on Combustion, Edinburgh, Scotland, July 3- Aug. 4, 2000.

Molina, A., E. G. Eddings, D.W. Pershing and A. F. Sarofim, “NO-Char Kinetics: Implications to NOx Emissions,” presentation at the ACERC Technical Conference on Addressing Regulatory Challenges in the Next Century, Salt Lake City, Utah (February 17-18, 2000).

Sarofim, A. F., E. G. Eddings and A. Molina, “Char Nitrogen Conversion: Implication to NO Emissions from Utility Boilers,” Plenary Lecture to be presented at the Mediterrean Combustion Symposium, June 20-25, 1999, Antalya, Turkey.

Zhang, H. and T. H. Fletcher, "Nitrogen Transformations during Secondary Coal Pyrolysis," poster presented at the 13th Annual ACERC Conference, Provo, Utah (February 25-26, 1999).

Zhang, H. and T. H. Fletcher, "Char Oxidation during Late Burnout," poster presentation at the 12th Annual ACERC Conference, Provo, Utah (March 25-26, 1998).

Molina, A., E. G. Eddings and A. F. Sarofim, “NOx Production During Pulverized Coal Combustion via the Char-N Route,” poster presentation at the 12th Annual ACERC Conference, Provo, Utah (March 25-26, 1998).

CURRENT AND PAST STUDENTS SUPPORTED UNDER THIS GRANT

- Alejandro Molina, Ph.D. student in Chemical Engineering, UofU
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